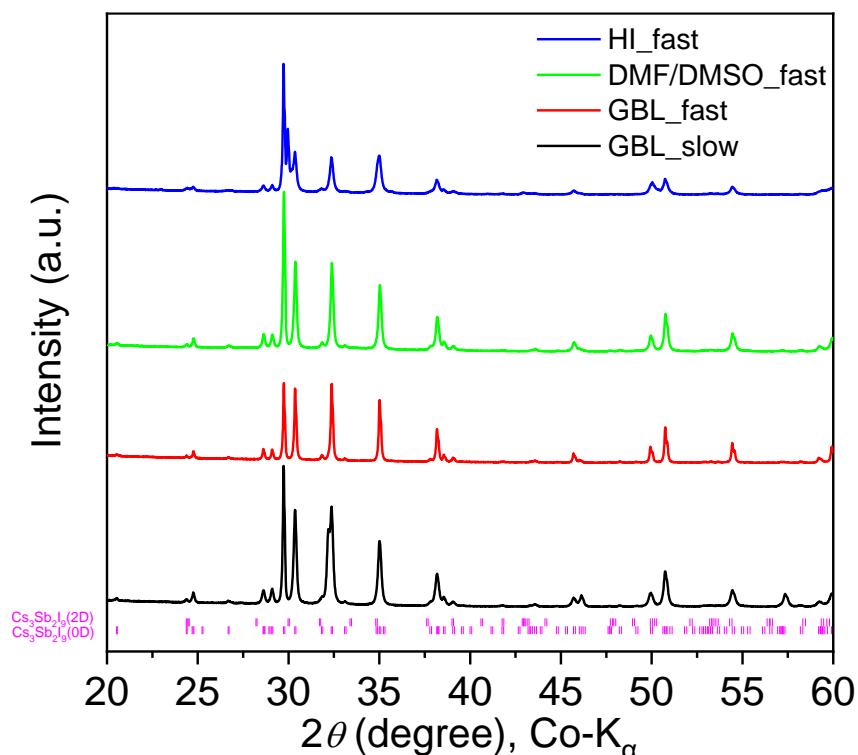


## Supporting Information

Tunable Optical and Thermochemical Properties of  $\text{Cs}_3\text{Sb}_2\text{I}_9$  Synthesized in Various Solvents

Samuel Meles Neguse, Songhak Yoon, Alexander Frebel, Dennis M. Jöckel, Marc Widenmeyer, Stefan Lange, Arnulf Rosspeintner, Stefan G. Ebbinghaus, Benjamin Balke, Anke Weidenkaff

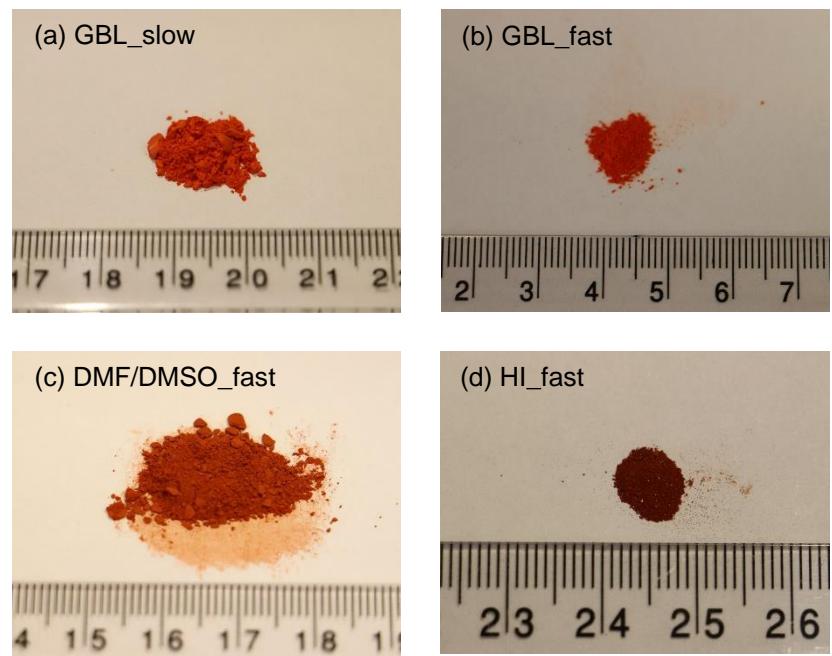
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**Figure S1.** An extended XRD patterns of GBL\_slow, GBL\_fast, DMF/DMSO\_fast, and HI\_fast. The tick marks at the bottom indicate the position of the Bragg reflections of either 2D or 0D  $\text{Cs}_3\text{Sb}_2\text{I}_9$ .

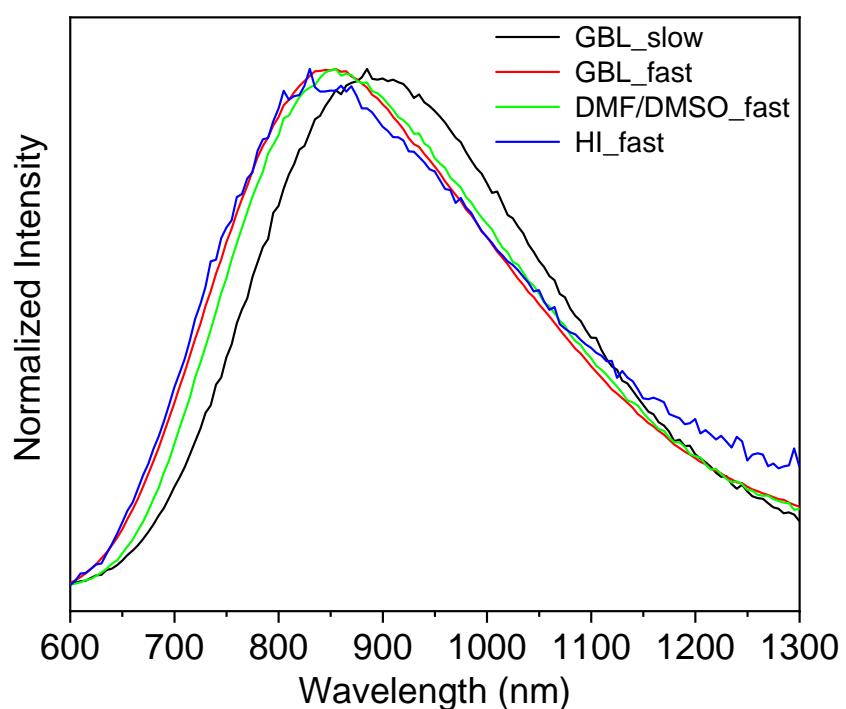
**Table S1.** Lattice parameters and unit cell volume determined by Le Bail fit of the powder X-ray diffraction patterns.

Sample	GBL_slow	GBL_fast	DMF/DMSO_fast	HI_fast
Lattice parameter $a$ ( $\text{\AA}$ )	8.3520(1)	8.3506 (1)	8.3485(1)	8.3521(2)
Lattice parameter $c$ ( $\text{\AA}$ )	20.9293(3)	20.9304(3)	20.9248(2)	20.9239(4)
$V(\text{\AA}^3)$	1264.35(3)	1263.97(3)	1263.00(2)	1264.05(4)

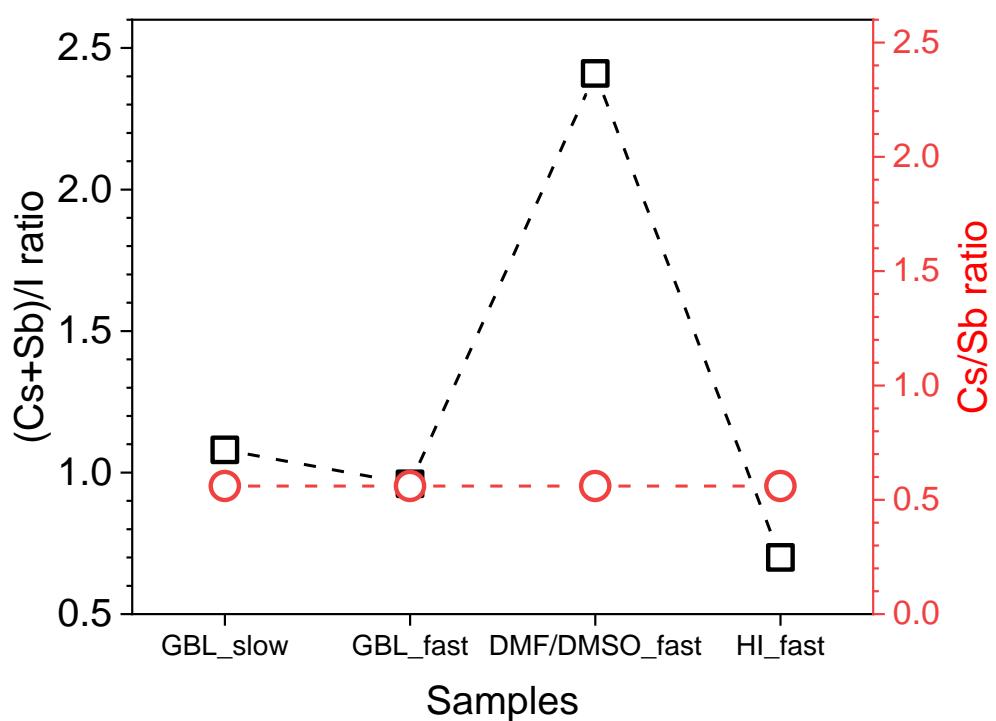
The numbers in parentheses are the estimated standard deviations.



**Figure S2.** Optical images of (a) GBL\_slow, (b) GBL\_fast, (c) DMF/DMSO\_fast, and (d) HI\_fast.



**Figure S3.** Photoluminescence emission spectra of  $\text{Cs}_3\text{Sb}_2\text{I}_9$  crystals ( $\lambda_{\text{exc}} = 440 \text{ nm}$ ) measured at room temperature in air.

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**Figure S4.** Ratios of  $(\text{Cs}+\text{Sb})/\text{I}$  in black (left) and the ratios of  $(\text{Cs}/\text{Sb})$  in red (right), obtained by XPS measurements.

**Table S2.** Summary of key findings.

	GBL_slow	GBL_fast	DMF/DMSO_fast	HI_fast
FWHM value of the Raman peak at $131\text{ cm}^{-1}$ ( $\text{cm}^{-1}$ )	7.54(38)	6.44(29)	7.19(35)	6.62(33)
Indirect bandgap (eV)	2.11	2.00	2.12	1.85
$E_U$ (meV)	118.2(15)	69.2(6)	158.8(8)	251.4(57)
PL emission peak (eV)	1.40	1.45	1.45	1.49
Carrier lifetime $\tau_1$ (ns)	1.05(1)	2.47(1)	2.23(2)	1.90(1)
Amplitude of the decay times $A_1$	0.997(7)	0.967(3)	0.921(1)	0.885(3)
Carrier lifetime $\tau_2$ (ns)	16.7(1.9)	301.3(20.6)	104.1(6.7)	19.9(5)
Amplitude of the decay times $A_2$	0.053(4)	0.034(1)	0.071(2)	0.151(2)
Average decay time $\tau_{average}$ (ns)	8.08(1.33)	244.4(19.8)	82.2(6.3)	13.44(42)

The numbers in parentheses are the estimated error ranges.